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Evaluation of an S2 Sampler for Receptor Modeling of Woodsmoke Emissions

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16. ABSTRACT

The Source Signature (S^2) sampler was developed to characterize the carbon (C) and elemental components in fine particulate emissions from a residential wood burner. The hot exhaust is sampled and diluted with filtered air to simulate normal diffusion. The resulting aerosol is passed through a 2.5-μm cyclone and the remaining particles collected on two parallel 1-μm filters, one quartz and one Teflon. The quartz filter allows analysis of carbon content and the Teflon filter allows elemental analysis by X-ray fluorescence. Both filters are also weighed to determine the mass-emission rate. The test program's main objectives were (1) to test and improve the performance of the method, and (2) to make a laboratory measurement of the elemental composition including the carbon component to establish a source signature for use in receptor modeling. The average cyclic emission rate measured during this test was 1 g of fine particulate per kg of wood burned. Main components of the particulate mass were: 35% volatile carbon, 6% elemental carbon, and 8% potassium (K).

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EVALUATION OF AN S^2 SAMPLER FOR RECEPTOR MODELING OF WOODSMOKE EMISSIONS

Introduction

Wood combustion emissions from residential woodstoves and their associated impact on air quality have recently been the subject of intensive research by a large body of investigators.[1] One area of research which is of interest to Federal, state, and local agencies concerned with air quality is the development of source receptor models[2] to calculate the impact of woodstove emissions on particulate mass loading present in the atmosphere.

Studies by Cooper and Maleks[3] and Courtney et al[4] suggest using the ratio of potassium to iron (Fe) in wood particulate (<2.5 μm aerodynamic diameter) to calculate the impact of wood combustion emissions on air quality. Wolff et al[5] have also used $^{12}C/^{14}C$ measurements, in addition to the K/Fe ratio, as a means of estimating the impact of wood combination emissions in Denver, CO, during a field study conducted in December, 1978. Muhlbaier et al[6] and the U.S. Environmental Protection Agency (EPA)[7] have studied emission factors for fireplaces and woodstoves. Muhlbaier et al[6] suggested that woodburning could account for 20 to 30% of the particulates during winter in Denver, CO.

At present a standard sampling procedure to collect emissions from woodstoves has not been developed. This paper describes the design and testing of a new sampling method for collecting fine particle emissions from woodstoves on Teflon and quartz filters. The aerosols collected on the Teflon filters are analyzed by X-ray fluorescence techniques[8] for elemental composition, and the quartz filters are analyzed by thermal combustion procedures[9] for volatile and non-volatile carbon content. Data are presented on elemental concentration and carbon composition in fine particles from the early, middle, and final stages of burning of pine, oak, poplar, and sweetgum. Comparisons are also made between the aerosol masses collected using the S^2 sampler and those collected using EPA Reference Method 5.[10]

Experimental Methods

S^2 Sampler

To establish a better understanding of the elemental signature of woodsmoke particulates, a sampler was developed for EPA with the capability to sample directly from a stack while simulating the normal dilution coalescence processes. The source signature sampler, S^2, has two parallel filter trains which collect equivalent particulate samples for X-ray and carbon analysis.

The primary design criteria for the S^2 sampler are:

1. The sampler must be portable to facilitate sampling from residential woodstoves in the field.

2. The sampler must collect equivalent samples of the particulate (< 2.5 μm) on two separate filters simultaneously. One filter should allow X-ray analysis of the sample and the other filter should allow carbon analysis of the sample.

3. The sampler must simulate the normal dilution and coalescence processes that occur as exhaust mixes with ambient air.

4. The raw exhaust should be isokinetically sampled.

5. The dilution chamber should be designed to have a Reynolds number of about 20,000 and to be at least ten diameters long to ensure adequate mixing.

6. The surface-to-volume ratio in the dilution chamber and probe lines should be as small as practical to limit surface losses and help prevent clogging.

Figure 1 is a diagram of the principal components of the S^2 sampler. The S^2 sample train is made up of an isokinetic sampling probe, a dilution system, the two parallel filter trains, and the necessary flow control systems. The raw wood smoke is sampled from the stack at approximately 20 lpm and diluted to approximately 200 lpm with clean air. This dilution takes place in a dilution tube, 25 cm long and 2 cm in diameter, having the necessary Reynolds number of about 20,000, at the flow rate of 200 lpm.

Equal portions of the diluted aerosols are pumped through the two filter trains. The particles larger than a 2.5 μm in aerodynamic diameter are cut from the stream in cyclones and designed to meet the specifications described by John et al.[11] The remaining fine particles are collected simultaneously on a Teflon (Ghia Corp.) and a quartz filter (Whatman Corp.). Both filters have a 1-μm pore size and are 37 mm in diameter. The Teflon filter is used in the X-ray analysis procedure.[8] The quartz filter is used in the carbon analysis procedure.[9] In addition to the S^2 sampler, this method requires measurement of the chimney or stack flow rate. Measuring this flow rate can be difficult, given the low velocity in natural draft chimneys, but a standard "J" type Pitot tube with an electronic manometer sensitive to 0.001 mm Hg provided suitable flow measurements in the laboratory.

Experimental Apparatus

The laboratory experimental apparatus used both for sampling method intercomparison experiments and for acquisition of filter samples for chemical analysis is shown schematically in Figure 2. A single airtight woodstone (Ashley model 7150-B) was used for all experiments. The stove was mounted on a floor balance capable of measuring weight differences of 250 g and the flue and damper assembly were flexibly coupled to the stove to permit periodic monitoring of combustion rate through weight loss.

In this test system, the flue gas is ducted into a horizontal, cylindrical, air-dilution apparatus similar to the type used for measuring automotive particles.[12,13] Flue gas is mixed with dilution air through an induction tube that faces downstream and terminates in the center of a mixing baffle. The combined flow rate of flue gas and dilution air is held constant at 12 m^3/min by the use of a pressure-recovery type of critical flow nozzle as a flow control element. The intake dilution air flow is also measured continuously by means of a laminar flow element. Thus, by looking at the difference in values between flow elements, the flue gas mass flow rate and the

dilution ratio can be calculated. For these experiments, the dilution ratio of air mass flow to flue gas mass flow was typically 6 to 1.

The air-dilution apparatus is also equipped with sampling ports for simultaneous recovery of the S^2 and Method 5 samples and with a large total particle collection device as well. The device includes a plate-and-frame filter assembly with pneumatically operated seals and accommodates a 0.5 x 0.5-m fluorocarbon-coated glass fiber filter (Pallflex T60A20) in a quick-change cassette.[14] The valving shown in Figure 2 permits temporary diversion of diluted flue gas during filter changing, a process which requires only a few seconds. Particle weight gain measured with this method was used as the reference estimate of particle mass emissions for comparison with S^2 on Method 5 measurements.

Sampling ports for stack gas samples were located in the exhaust flue about 1.5 m above the stove exhaust as shown in Figure 2. Flow control for the S^2 and Method 5 samplers was handled manually during the burn period. In practice, the stack gas flow rate was highly variable both during a burn and from run-to-run. Therefore, it was difficult to maintain isokinetic flow in the stack gas samplers. This flow variability also has significance in measurement of emissions factors taken over a burn period. Since neither the exhaust flow nor the emission factor is constant over a burn, it is difficult to achieve a constant sample to flue gas ratio and, thus, correctly proportion each element of the burn in a composite sample. Generally, this flow control difficulty could have caused an uncertainty in the Method 5 or the S^2 flue gas measurements of ±20%.

Chemical analysis of sampled particles was conducted by standard methods. Elemental analysis was done by X-ray fluorescence analysis using a multichannel wavelength dispersive instrument. Elemental and volatile carbon were measured by a previously reported combustion technique [Stevens et al, 1982].

Test Description

During the test program, several different fuels were burned in the Ashley wood stove. Oak, pine, poplar, and sweetgum were tested. All of the fuel had been dried to approximately 10% moisture content except that in addition to the dry oak, green and partially seasoned oak were also tested. The green oak had a 50% moisture content and the partially seasoned oak had about 30% moisture content. (These moisture contents were measured in the wood fuel at the time of the tests with a Delmhorst model RC-1 moisture meter.)

From each burning load of fuel, several samples were taken--one at the early stage, two at the middle stage, and one at the late or charcoal stage of the fire. The different samples were taken to test the performance of the S^2 at several different emission levels. The samples also provided elemental composition data for the various stages of burning. Figure 3 shows idealized test conditions for this group of tests. Only approximations of these conditions are found in reality. For test purposes, the burn stage was defined largely by stack temperature and by observation of the fire by the person conducting the test.

The early stage of burning is characterized by rising temperatures; drying and revolatilization are the dominant burning processes. The middle stage is characterized by relatively constant temperatures and burn rates; pyrolysis is the dominant burning process. The last stage is characterized by slowly decaying temperatures; charcoal burning is dominant.

Comparison between the S^2 Sampler, Method 5, and the Total Particulate Filter

Figure 4 is a graphic comparison of the particulate emission levels measured by S^2 and Method 5; these together are compared to the control filter's "total load" sample. During this test program 42 tests were made. The emission levels were usually about 1 or 2 g/kg of particulates to dry fuel and it was only during the early stages of the burn cycle that significantly different emission levels were observed. Taking the total particulate filter data as the best emission rates, there is a spread in the emission levels of 2 orders of magnitude. So the results showing a more or less linear relationship between the three different methods are a good indication that the S^2 sampler takes adequately representative samples. Table 1 is a summary of the comparison between the emission levels measured by S^2, Method 5, and the control filter. Given here are the cyclic emission levels integrated over the burn cycle. Although the average emission levels had large standard deviations, the different methods agreed to within 30% of each other.

The mean of the ratio of S^2 emission levels to Method 5 levels was 0.70 and the mean of the S^2 emission levels to the control filter emission levels 0.80. One reason that the S^2 numbers were lower is that they represent the emission levels of the fine particulate, whereas the other two methods represent the total load. The large standard deviations were due to the fact that many burn parameters had not been controlled.

Table 2 presents the results of the S^2 sampler in terms of the flow performance. Problems with the flow control caused the flow parameters to differ by a factor of 3. However, since the particles were $< 2\ \mu m$, this non-isokinetic and non-proportional sampling should not have affected the emission level numbers significantly because the actual variation in the emission levels is large ($\sim$ 2 orders of magnitude).

An effort was made to determine what the particulate losses were in the S^2 sample train. The sampler probe and dilution system was disassembled at the end of the tests and the segments washed down with acetone. The procedure is similar to the probe wash procedures for Method 5. Figure 5 shows the S^2 sampling train with data in percent of total particulate load. The data show approximately 25% of the particulate material being collected in the probe tip, transfer line, and mixing nozzle. The balance of the material was collected on the filter media. Although the line and probe were as short as possible under the circumstances, these data indicate that the probe may need to be heated if operated in a cold environment.

Elemental Data

Table 3 summarizes the elemental analysis of the filters collected in this laboratory test program. Included on the chart are the averages

Table 1. Comparison of S^2, Method 5, and total particulate filter sampler by emission rates and cyclic averages.

Sampler	Mean Emissions Levels (g/kg)	Std. Devs.‡‡ (g/kg)	No. Obvs.
S^2*			
• Teflon	1.02	0.99	28
• Quartz	0.95	0.90	28
• S^2 and probe wash	1.45	—	1
EPA—Method 5**			
• Filter only	1.28	0.93	28
• With probe wash	1.79	1.23	28
Total particulate filter‡	1.17	1.14	28

*Particulates <2 μm.
**Particulates and condensables above 390 K.
‡Particulates and condensables above 310 K.
‡‡These numbers reflect the variations in the actual emission levels from test to test and from the different stages of the burn cycle. The uncertainty in each observation of the three methods is estimated to be:

S^2 ~ 20%
Method 5 ~ 20%
Total particulate filter ~ 10%

Table 2. S^2 sampler flow performance.*

Parameters	Means	Std. Devs.	No. Obvs.
Dilution ratio $\left(\frac{\text{total flow}}{\text{raw exhaust}}\right)$	3.0	0.9	42
% isokinetic $\left(\frac{\text{probe tip velocity}}{\text{stack velocity}}\right)$*100%	360%	140%	42
Total flow in dilution tube	124 lpm	12 lpm	42
Raw exhaust sample rate	40 lpm	12 lpm	42
Reynolds number	6.96×10^3	6.74×10^2	42

*All flows are at 20° C and 760-mm Hg.

Table 3. Summary of elemental analysis (percent particulate mass).

No. Obvs.	Fuel/ Moisture (%)	Burn	Par.*	Sulfur	Chlorine	Potassium	Volatile Carbon	Elemental Carbon	Total Detected	Emission Rate (g/min)
1	Pine, 10%	E	$\bar{P}$**	0.0	0.1	0.0	44.5	2.1	46.8	1.227
			σ‡	—‡‡	—	—	—	—	—	—
4		M	$\bar{P}$	0.2	0.1	0.6	30.5	8.5	43.7	0.194
			σ	0.2	0.1	0.3	10.4	5.8	8.7	—
1		L	$\bar{P}$	1.2	0.5	29.5	16.9	0.0	48.7	0.026
			σ	—	—	—	—	—	—	—
		integrated		0.1	0.1	1.0	38.7	4.5	45.7	0.299
3	Poplar, 10%	E	$\bar{P}$	2.8	0.3	12.5	33.1	9.5	58.6	0.218
			σ	3.6	0.1	17.7	18.4	8.6	7.3	—
3		M	$\bar{P}$	5.5	0.8	21.1	38.9	4.8	71.5	0.033
			σ	2.2	0.6	2.9	3.2	1.7	8.1	—
3		L	$\bar{P}$	1.9	0.3	27.5	14.6	0.7	45.2	0.019
			σ	1.1	0.3	3.8	4.3	0.8	3.2	—
		integrated		3.6	0.5	16.9	33.2	7.0	61.6	0.055
1	Sweet gum, 10%	E	$\bar{P}$	1.0	0.3	2.8	47.6	10.5	62.6	0.090
			σ	—	—	—	—	—	—	—
3		M	$\bar{P}$	5.8	0.5	18.1	27.2	5.9	58.9	0.022
			σ	4.4	0.2	14.9	2.0	2.4	16.6	—
3		L	$\bar{P}$	5.2	0.6	23.6	18.1	11.1	60.2	0.023
			σ	3.4	0.4	14.8	15.3	14.5	12.9	—
		integrated		3.9	0.4	13.5	33.0	8.6	60.5	0.031
2	Oak, 10%	E	$\bar{P}$	0.1	0.4	0.1	30.5	5.6	36.9	0.456
			σ	0.0	0.3	0.1	7.5	0.9	8.0	—
6		M	$\bar{P}$	3.9	4.3	15.5	30.1	11.1	67.2	0.076
			σ	3.1	4.5	11.5	19.0	11.3	27.6	—
2		L	$\bar{P}$	1.6	0.5	32.3	14.3	0.0	49.2	0.024
			σ	1.1	0.2	2.6	10.7	0.0	6.7	—
		integrated		1.6	1.8	7.7	29.2	7.2	48.5	0.115
2	Oak, 30%	E	$\bar{P}$	0.1	0.1	0.8	44.5	1.8	47.5	0.581
			σ	0.1	0.0	0.4	1.1	0.6	0.1	—
2		M	$\bar{P}$	1.1	0.8	5.2	44.7	0.9	52.9	0.079
			σ	0.4	0.1	2.1	4.0	0.9	5.6	—
1		L	$\bar{P}$	0.8	0.3	13.6	49.1	4.0	68.4	0.008
			σ	—	—	—	—	—	—	—
		integrated		0.2	0.2	1.5	44.5	1.6	48.3	0.249
1	Oak, 50%	E	$\bar{P}$	0.3	0.2	1.7	41.5	4.5	48.3	0.208
			σ	—	—	—	—	—	—	—
1		M	$\bar{P}$	3.2	1.3	13.6	45.1	5.9	59.3	0.038
			σ	—	—	—	—	—	—	—
2		L	$\bar{P}$	2.2	1.2	15.1	42.2	3.7	65.8	0.001
			σ	0.8	0.9	4.9	3.7	0.6	4.7	—
		integrated		1.5	0.7	6.8	43.0	5.1	53.0	0.045
10	Average all fuel	E	$\bar{P}$	1.0	0.3	4.4	38.3	6.0	50.2	0.480
			σ	2.1	0.2	10.0	11.3	5.4	10.1	—
19		M	$\bar{P}$	3.3	1.7	12.5	33.4	7.4	59.7	0.080
			σ	3.2	3.0	11.0	12.6	7.3	19.2	—
12		L	$\bar{P}$	2.6	0.6	24.3	23.1	3.9	55.3	0.020
			σ	2.3	0.5	9.4	15.1	7.7	10.9	—
		integrated		1.8	0.8	8.0	35.9	6.4	53.6	0.130

* Parameter.

** $\bar{P}$ = mean % particulate.

‡ σ = standard deviation.

‡‡ — = Only one observation; standard deviation does not apply.

from the analysis of several different filters loaded during similar burn conditions. Also included are the standard deviations of each group. The carbon data were derived from carbon analysis of the quartz filters. The total carbon content is further subdivided into two components: volatile carbon and elemental carbon. The volatile component is defined operationally as the portion of the particulate which pyrolyzes at 630° C in a helium atmosphere. The elemental component is defined operationally as the part of the particulate which remains after volatile analysis, and which undergoes combustion at 850° C in a 98% helium and 2% oxygen atmosphere. The Teflon filters were analyzed using X-ray fluorescence techniques.

The total carbon content of the particulates collected during this study was typically 42% of the total particulate mass. The elemental carbon portion was measured to be about 6%, and the volatile carbon was measured to make about 36% of this mass.

Potassium, the next largest component, was measured to be about 8% of the total particulate. The carbons and K together account for about 50% of the particulate mass. The remaining 50% may be oxygen which makes up approximately 40% of the dry fuel weight.

Figure 6 shows the elemental composition of wood smoke particulate as it varies between stages of burning. From this chart the percent of potassium appears to increase during the burn cycle while the percent of carbon compounds appears to decrease. These changes in chemical make-up may reflect different burning processes taking place during the different stages. Or the different percents of elements may be due to difference in combustion temperatures between K and C. The ratio of K to total C varies from about 0.1 to 0.9 during the cycle with an average for all fuel tested of 0.2.

Comparisons in the composition of particulate during middle stage of burning of different woods are depicted in Figure 7. The only noticeable differences appear to be between the dry pine and the other fuels. These numbers are the combined results of four separate pine fuel tests, and should be considered only as preliminary data. Additional work is needed to clarify these values.

The variations in elemental content of woodsmoke particulate as correlated with fuel moisture content are shown in Figure 8. These values were calculated from filters loaded during the middle stage of burning oak having three moisture contents: 10%, 30%, and 50%.

The "T bars" on these figures reflect actual test to test and cycle to cycle variations in the elemental composition of the particulates. The uncertainty in the X-ray fluorescence analysis was 0.01% of particulate mass. The uncertainty in the total carbon analysis was 7% of particulate mass.

Conclusions

1) The S^2 sampler takes a reasonably representative sample from the stack.

2) Problems with the S^2 flow controller need to be corrected to allow the gas to be diluted by at least 10 to 1 and to allow a better proportional sample to be taken. However, the actual variations in the emission rates seem to hide this uncertainty.

3) The relative amounts of potassium and of total carbon in the collected particulates change during the different burn stages. Percent K increases during the middle and final stages; percent total C decreases during the middle and final stages. The average ratios of K/C for the burn cycle of the following wood types were pine, 0.02, poplar, 0.42, sweetgum, 0.32, and oak, 0.21.

4) No significant differences were noted in the composition of the particulate between different woods or different fuel moisture contents except for pine. Pine had a significantly lower potassium content than the other fuels. Because few pine samples were collected, further study of this wood is recommended before any conclusions are drawn.

5) An average of 1 g of fine particulate was collected per kg of dry wood burned (all cases). The analysis of these particles indicates that 0.36 g/kg of volatile carbon, 0.06 g/kg of elemental carbon, and 0.08 g/kg of potassium were present. Sulfur, chlorine, and other trace elements detected comprise an additional 0.04 g/kg of the particulate emissions per kg of fuel. The remaining 0.46 g/kg of particulate mass is thought to be largely oxygen since oxygen makes up about 40% of the mass of the original fuel. In these tests, the average burn rate was 9 kg/hr and the average mass of wood burned as 16 kg.

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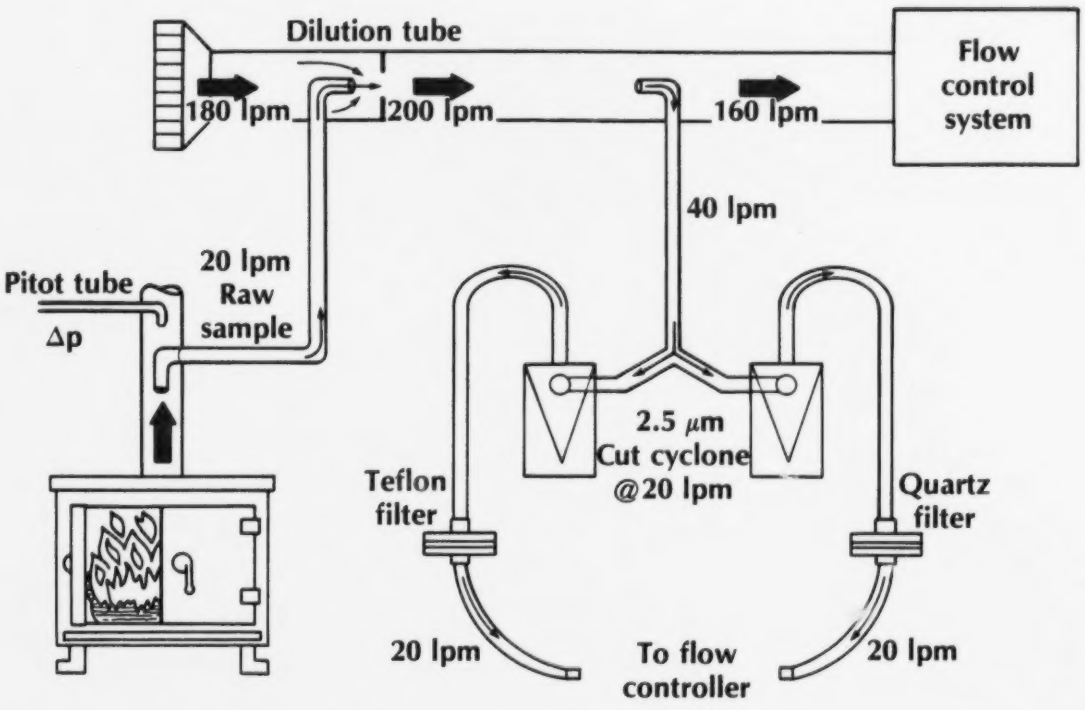


Figure 1. S^2 sample flow.

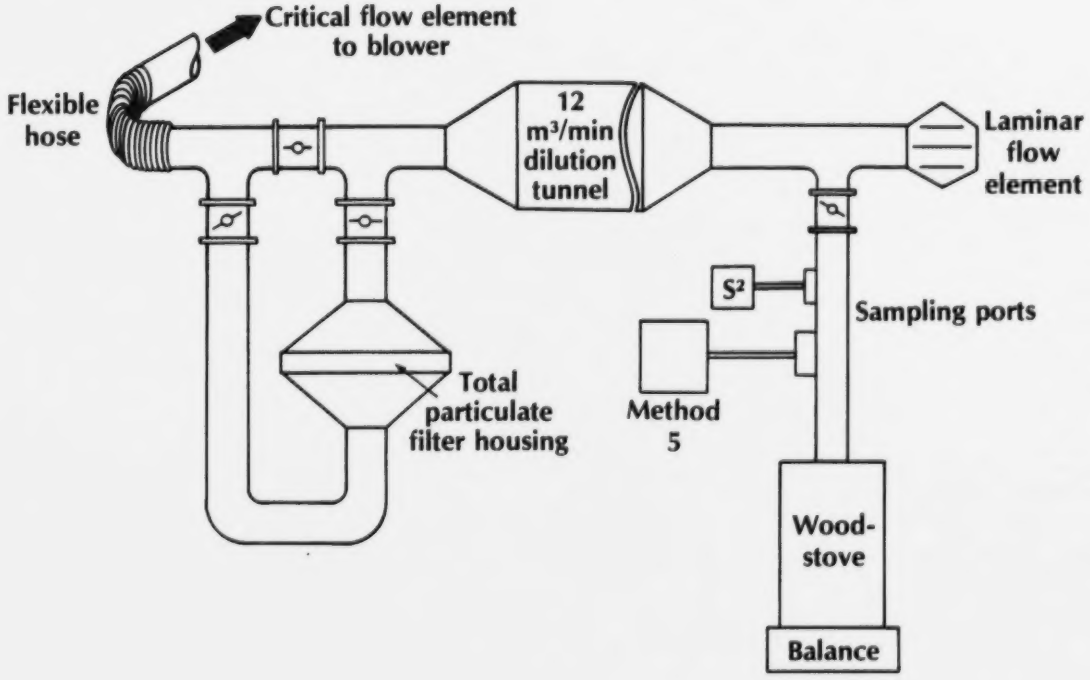
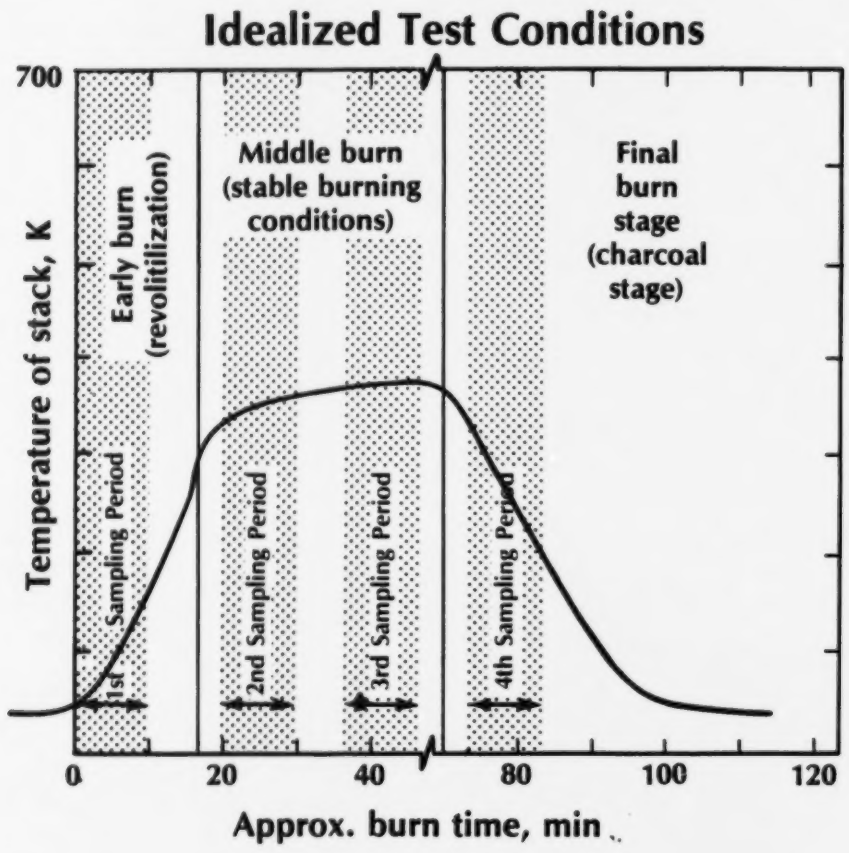


Figure 2. Laboratory experimental apparatus.



Test Conditions	Mean	Std. Devs.	No. Obvs.
Average burn rate	9.1 kg/hr	3.2 kg/hr	6
Average load of fuel	16.1 kg	3.5 kg	6
Average burn time	95 min	25 min	6
Stack temperature	426 K	76 K	42
Stack velocity	2.8 mps	0.7 mps	27
Stack flow rate*	2.0 cmm	0.4 cmm	27

*Flow reported at 20°C, 760 mm Hg.

Figure 3. Idealized test conditions for tests demonstrating different burning stages and mean values of test parameters.

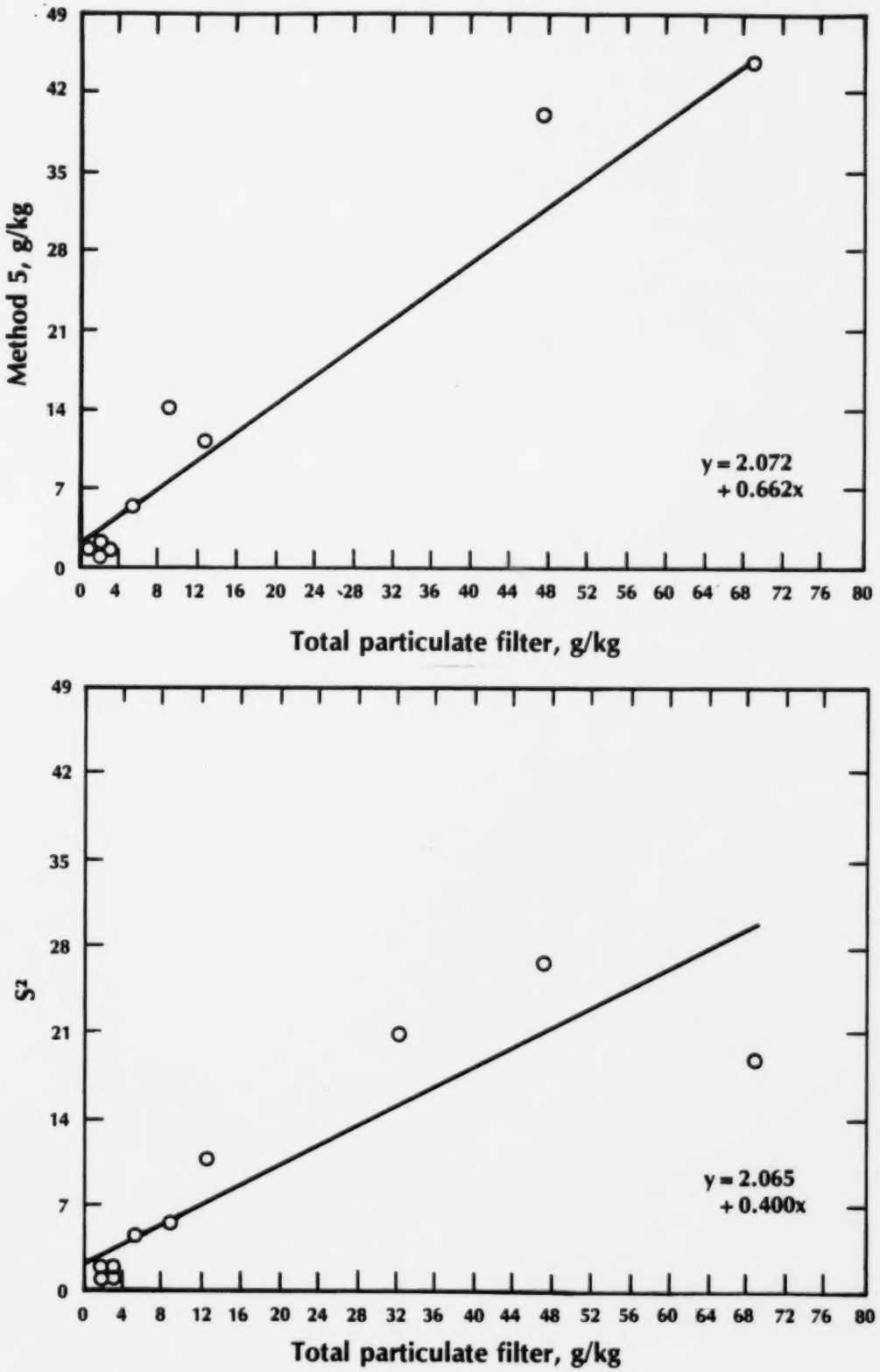


Figure 4. Graph of particulate emission levels using S^2, Method 5, and the total particulate filter.

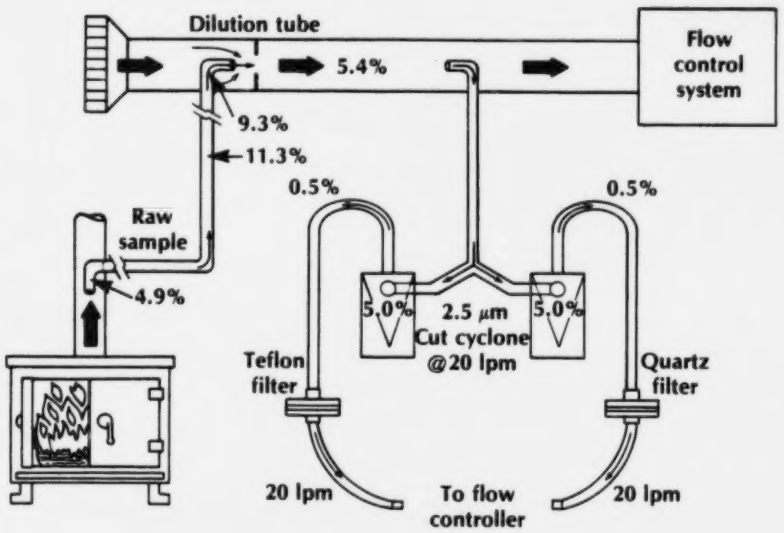


Figure 5. S^2 sampling train; percent of total load sampled.

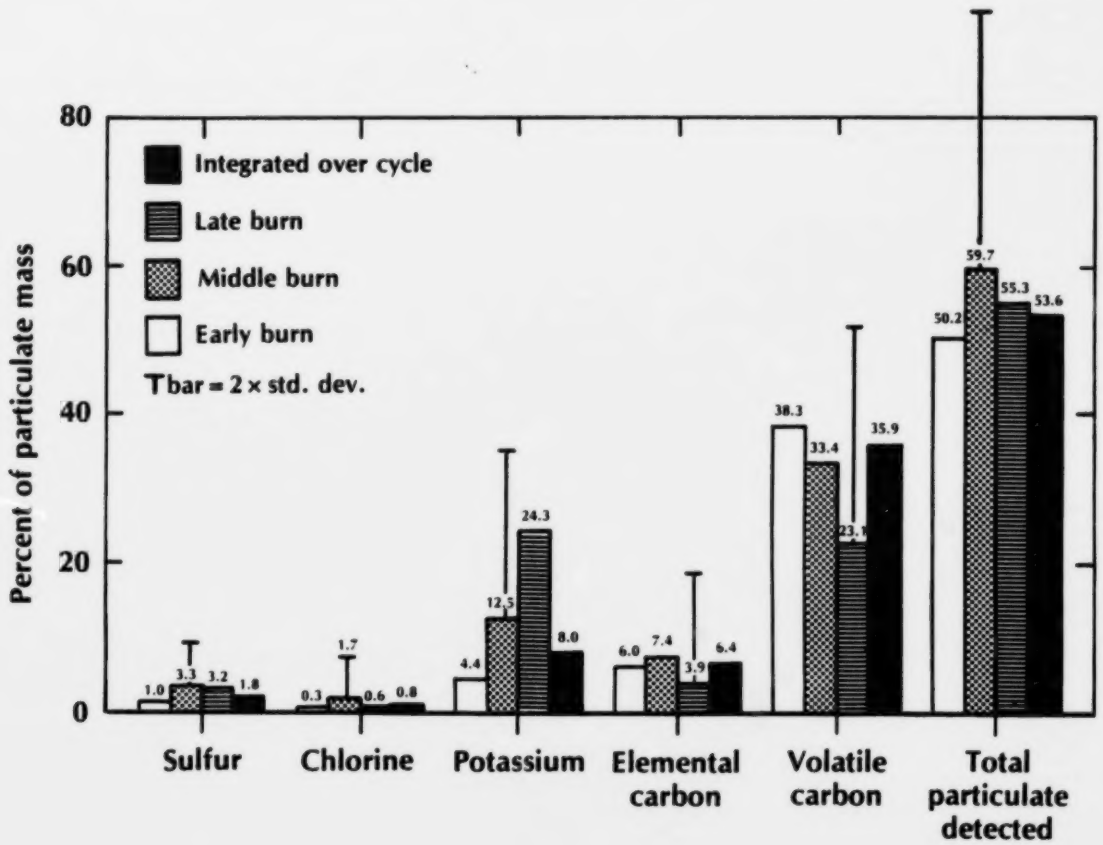


Figure 6. Variations in elemental composition of woodstove emissions during different burning stages.

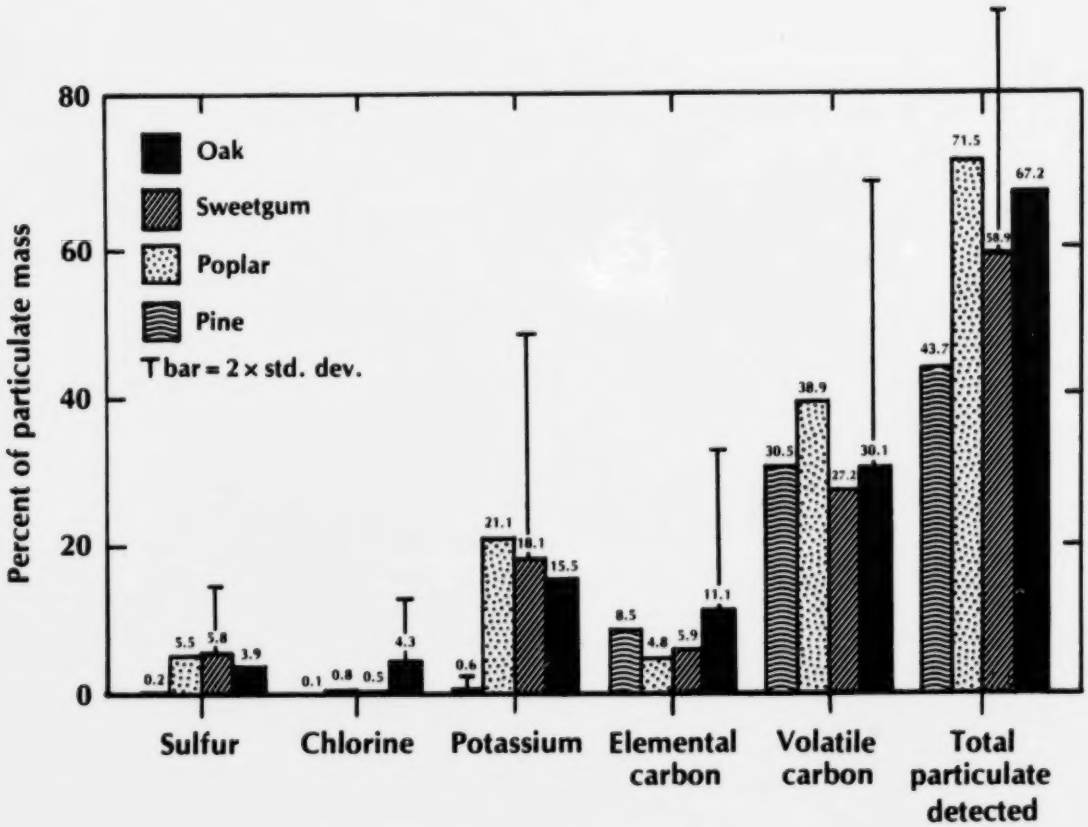


Figure 7. Comparison of the particulate composition of oak, sweetgum, poplar, and pine during the middle stage of burning.

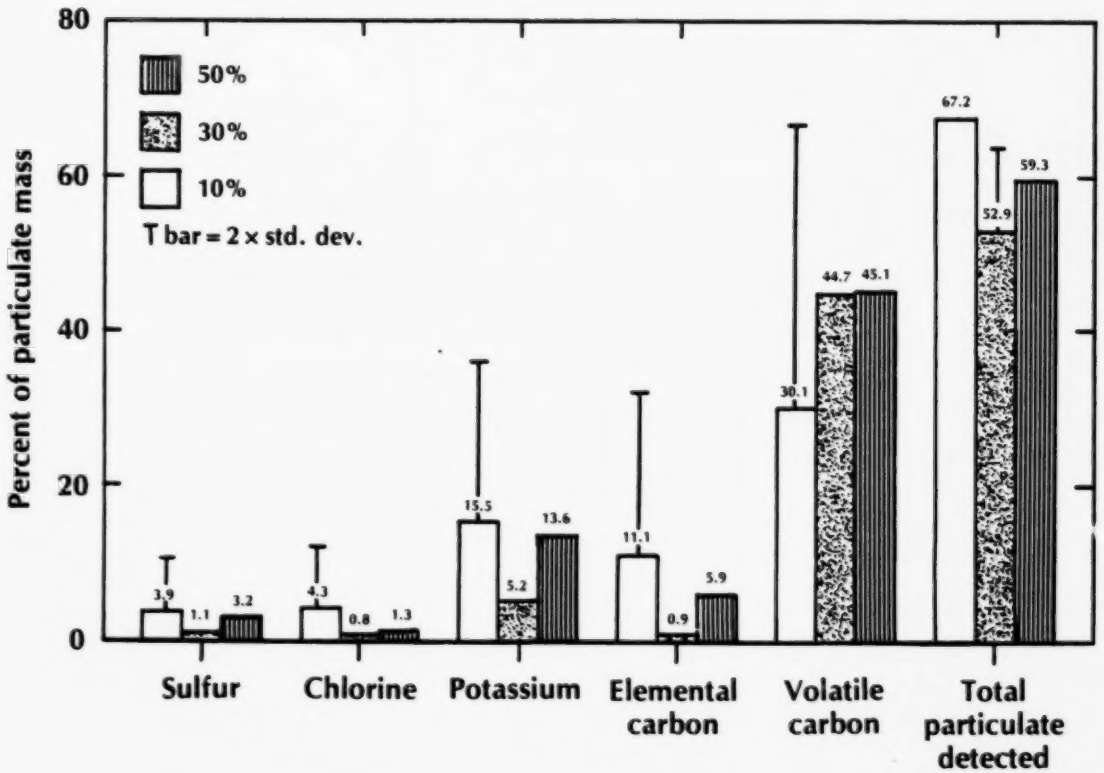


Figure 8. Elemental composition of woodsmoke particulate as correlated with fuel moisture content.